

# Filling-Factor-Dependent Magnetophonon Resonance with Circularly Polarized Phonons in Graphene Revealed by High-Field Magneto-Raman Spectroscopy

Y. Kim,<sup>1</sup> J. M. Poumirol,<sup>1</sup> A. Lombardo,<sup>2</sup> N. G. Kalugin,<sup>3</sup> T. Georgiou,<sup>4</sup> Y. J. Kim,<sup>4</sup>  
K. S. Novoselov,<sup>4</sup> A. C. Ferrari,<sup>2</sup> J. Kono,<sup>5</sup> O. Kashuba,<sup>6</sup> V. I. Fal'ko,<sup>7,8</sup> and D. Smirnov<sup>1,\*</sup>

<sup>1</sup>*National High Magnetic Field Laboratory, Tallahassee, FL 32310, USA*

<sup>2</sup>*Engineering Department, Cambridge University, Cambridge, CB3 0FA, UK*

<sup>3</sup>*Department of Materials and Metallurgical Engineering, New Mexico Tech, Socorro, NM 87801, USA*

<sup>4</sup>*School of Physics & Astronomy, University of Manchester, Oxford Road, Manchester M13 9PL, UK*

<sup>5</sup>*Department of Electrical and Computer Engineering and Department  
of Physics and Astronomy, Rice University, Houston, TX 77005, USA*

<sup>6</sup>*Institute for Theoretical Physics A, RWTH Aachen, D-52074 Aachen, Germany*

<sup>7</sup>*Department of Physics, Lancaster University, Lancaster, LA1 4YB, UK*

<sup>8</sup>*DPMC, Université de Genève, CH-1211 Genève 4, Switzerland*

(Dated: November 27, 2012)

We perform polarization-resolved Raman spectroscopy on graphene in magnetic fields up to 45T. This reveals a filling-factor-dependent, multi-component anti-crossing structure of the Raman G peak, resulting from magneto-phonon resonances between magneto-excitons and circularly polarized  $E_{2g}$  phonons. This structure is explained with a model of Raman scattering taking into account the effects of spatially inhomogeneous carrier densities and strain. This shows that random fluctuations of strain-induced pseudo-magnetic fields lead to increased scattering intensity inside the anti-crossing gap, consistent with the experiment.

The magneto-phonon resonances (MPR) effect, observed in semiconductors when the energy of an optical phonon coincides with the inter-Landau level (LL) separation [1], is an important tool for the investigation of electron-phonon interactions, especially in two dimensional systems[2]. Electron-phonon coupling (EPC) in graphene and graphite has been studied for several years[3–7]. The zone-centre, doubly degenerate,  $E_{2g}$  phonon strongly interacts with electrons, resulting in renormalization of phonon frequencies and line broadening[8–13]. These are tunable by electric and magnetic fields, through Fermi-energy shifts and Landau quantization. The Raman G peak in graphene is predicted to exhibit anti-crossings when the  $E_{2g}$  phonon energy matches the separation of two LLs[14–16]. This MPR effect can be described as a resonant mixing of electronic and lattice excitations into a combined mode, leading to a splitting proportional to the EPC[15], and was observed in magneto-Raman scattering on SLG on the surface of graphite [17, 18], non-Bernal stacked multilayer graphene on SiC[19], and bulk graphite[20].

Here, we report a polarization-resolved Raman spectroscopy study of MPR in single layer graphene (SLG), demonstrating a strong dependence of the MPR line-shape on the Raman polarization and carrier density. This is explained as a manifestation of MPR between electronic magneto-excitons and circularly polarized optical phonons[15], combined with the effect of inhomogeneous carrier densities and strain.

The energy spectrum of SLG in a perpendicular magnetic field  $B$  consists of discrete fourfold (spin and valley) degenerate Landau Levels (LL) with energies  $E_n = \text{sign}(n)\sqrt{2|n|\hbar\tilde{c}/l_B}$ , where  $n = \dots, -2, -1, 0, 1, 2, \dots$  is the

index of LLs in the conduction ( $n>0$ ) and valence ( $n<0$ ) bands,  $n = 0$  being exactly at the Dirac point (see Ref.21 and references therein). The band velocity,  $\tilde{c}$ , is the slope of the Dirac cone at zero  $B$ , and  $l_B = \sqrt{\hbar/eB}$  is the magnetic length. The LL occupancy is characterized by the filling factor  $\nu = 2\pi l_B^2 \rho_s$ , where  $\rho_s$  is the carrier density. Fully filled LLs with  $n = 0, 1, \dots$  have  $\nu = 2, 6, \dots$ [21]. MPR in graphene occurs when the energy of the inter-band inter-LL transitions,  $-n \rightarrow n \pm 1$ , matches that of the  $E_{2g}$  phonons, resulting in strongly coupled electron-phonon modes, in which the electronic magneto-exciton is an anti-symmetric superposition of inter-LL excitations in each of the two SLG valleys, K and K', while this gives symmetric superposition active in far-infrared absorption[15, 22]. This effect is strongest for the  $E_{2g}$  MPR with inter-Landau-level transitions  $-1 \rightarrow 0$  and  $0 \rightarrow 1$ , at  $B_{MPR}^0 \approx (25 - 30)\text{T}$ . Since the symmetry of such electronic excitation[22], as well as of the  $E_{2g}$  phonons, allows them to be active in Raman scattering, MPRs manifest through a fine structure which develops in the Raman G peak in the vicinity of the MPR condition,  $\Omega_\Gamma = [\sqrt{|n|} + \sqrt{|n|+1}]\sqrt{2\hbar\tilde{c}/l_B}$  at  $B_{MPR}^n$ .

A specific feature of MPR in SLG, is the  $\nu$  dependence of the anticrossing fine structure of coupled electron and phonon modes (which can be tuned externally by varying the carrier density), as well as a unique possibility to resolve the MPR of circularly polarized phonons[15] (Fig.1). The polarization properties of Raman scattering involving magnetoexcitons are such that the incoming and outgoing photons have opposite circular polarizations, with angular momentum transfer  $\pm 2\hbar$ , of which  $\pm 3\hbar$  is coherently transferred to the lattice, while the remaining angular momentum transfer  $\mp 1\hbar$  goes into elec-

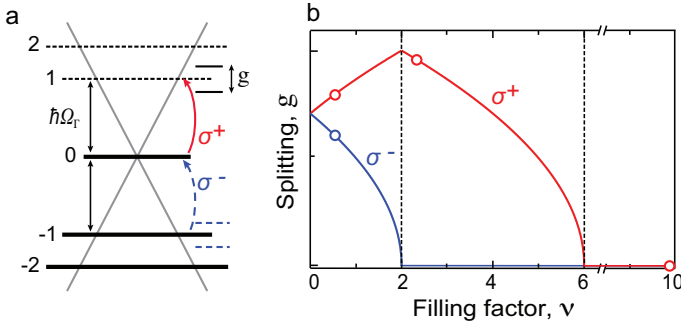


FIG. 1. (a) (grey lines) SLG band structure at  $B=0$ . In the vicinity of MPR between  $0 \rightarrow 1$  electron-hole excitations and  $E_{2g}$  phonon, at  $B_{MPR}^0 \approx (25-30)\text{T}$ , mode splitting, indicated by  $g$ , ensues. (b) Calculated[15] splitting as a function of filling factor,  $\nu$ . Open circles indicate  $\nu$  probed in our experiment

tronic excitations[22]. In the following, we will refer to this angular momentum transfer as a  $\sigma^\mp$ -polarized transition between the electronic LLs, with opposite signs for  $-n \rightarrow n \pm 1$  inter-LL excitons:  $\sigma^+$  for  $-n \rightarrow n+1$  and  $\sigma^-$  for  $-n-1 \rightarrow n$  valence-to-conduction band transitions. Blocking such transitions by filling (depleting) the final states or depleting (filling) the initial states with electrons would suppress (promote) coupling between magneto-excitons and phonons. Since the  $\sigma^+$  and  $\sigma^-$  magneto-excitons are based on different LL pairs, changing carrier density in SLG would affect differently the size of the anticrossing in the MPR of  $\sigma^\mp$  circularly polarized phonons, as illustrated in Fig.1. At  $\nu = 0$ , corresponding exactly to a half-filled  $n = 0$  LL, the coupling strength of  $\sigma^+$ - and  $\sigma^-$ -polarized modes is equal, causing the G peak to split equally for  $\sigma^+$  and  $\sigma^-$  phonon polarizations[15]. For  $0 < \nu < 2$ , corresponding to a partially filled  $n = 0$  LL, the  $-1 \rightarrow 0$  transition becomes partly blocked, while the  $0 \rightarrow +1$  transition is promoted, giving rise to different splittings in the fine structure of  $\sigma^+$ -polarized and  $\sigma^-$  polarized modes. For  $2 < \nu < 6$ ,  $n = 0$  LL is full, leaving no space for MPR with the  $\sigma^-$  phonon, whereas  $n = +1$  LL is only partly filled, resulting in a MPR-induced fine structure in the  $\sigma^+$  phonon lineshape (with maximum splitting at  $\nu = 2$ ). Thus, the EPC at the MPR resolves energies of the  $\sigma^\mp$ -polarized lattice excitations[15, 22]. Note that, in order to probe the direct B influence on the motion of lattice nuclei would require unattainably strong  $B \sim 10^4\text{T}$  or higher, as can be estimated from the carbon-atom-to-electron mass ratio. Finally, at high  $\nu > 6$ , both  $-1 \rightarrow 0$  and  $0 \rightarrow 1$  transitions are blocked, completely suppressing the MPR-induced fine structure for both modes.

We measure a SLG on Si/SiO<sub>2</sub> prepared as follows. Large area graphene films are grown on copper foils by chemical vapor deposition (CVD)[23, 24]. A  $\sim 25\mu\text{m}$  thick piece of Cu foil is initially cleaned with acetone, deionized (DI) water, and isopropanol to remove organic and

inorganic contaminants from the surface. The foil is then loaded in a 4 inch quartz tube, and annealed at  $1000^\circ\text{C}$  for 30mins with an H<sub>2</sub> flow of 20 cubic centimeters per minute (sccm) and a pressure of 200mTorr. Such annealing process increases Cu grain size and removes native oxide layer. For the growth of graphene films, a gas mixture of H<sub>2</sub> and CH<sub>4</sub> (flow rates of 40 and 20sccm respectively) is injected in the CVD chamber for 30mins while maintaining the pressure at 600mTorr and temperature at  $1000^\circ\text{C}$ . Finally, the sample is rapidly cooled to room temperature under a hydrogen flow at a pressure of 200mTorr. Such procedure results in high quality SLG on both sides of the foil, as shown by Raman spectroscopy[4, 25]. A layer of Poly(methyl methacrylate) (PMMA) is then spun on one of the surfaces of the Cu foil. The graphene layer on the other side of Cu is removed with an oxygen plasma at a pressure of 20mTorr and a power of 10W for 30s. The PMMA/graphene membrane is then released by dissolving the Cu in a 0.2M aqueous solution of ammonium persulphate. The remaining PMMA/graphene layer is cleaned several times by DI water to remove residual salt, and transferred onto the SiO<sub>2</sub>/Si substrates. After the sample is dried in ambient conditions, the PMMA is removed in acetone, leaving the graphene adhered to SiO<sub>2</sub>/Si[24]. Using the dependence of graphene Raman spectra upon doping, the carrier density is estimated by combining the intensity and area ratio of the G and 2D peaks,  $I(2D)/I(G)$  and  $A(2D)/A(G)$ , with the position and Full Width at Half Maximum of these peaks,  $\text{Pos}(G)$ ,  $\text{Pos}(2D)$ ,  $\text{FWHM}(G)$ ,  $\text{FWHM}(2D)$ [7, 8, 26]. Under ambient laboratory conditions, the SLG sample is initially p-doped with a carrier concentration  $\sim 5 \times 10^{12}\text{cm}^{-2}$ . By adjusting annealing parameters (typically 8 hours in 90%/10% Ar/H<sub>2</sub> atmosphere at temperatures up to  $220^\circ\text{C}$ ) and degassing in  $< 10^{-4}\text{mbar}$  vacuum, the sample can be rendered n-type, with an electron density  $\sim 2 \times 10^{12}\text{cm}^{-2}$ . Exposing the sample to low-pressure ( $\sim 1\text{mbar}$  or less) N<sub>2</sub> atmosphere reduces the carrier density to  $\sim 0.4 \times 10^{12}\text{cm}^{-2}$ . The exposure to ambient pressure N<sub>2</sub> gas or air results in hole doping, restoring p-type doping with carrier densities  $\sim (5-6) \times 10^{12}\text{cm}^{-2}$ . If the sample is left in ambient air for a long period of time, it continues to experience p-doping reaching a carrier concentration  $\geq 10^{13}\text{cm}^{-2}$  in several weeks. However, this doping is inhomogeneous, with a 10 to 20% variation, as shown by Raman mapping.

B dependent Raman measurements are performed in a quasi-backscattering configuration at 300K for B up to 45T using a high-field magneto-optical insert, as for Ref.20. The combination of a linear polarizer and a  $\lambda/4$  plate, circularly polarizes the incident and scattered light. The excitation beam (532nm) is focused to a spot  $\lesssim 10\mu\text{m}$ , with a power  $\sim 4\text{mW}$ . The circularly polarized  $\sigma^\pm$  excitation, probed using the  $\sigma^\pm/\sigma^\mp$  configurations of out/in polarisations, is achieved by reversing the B polarity. In this notation, the first (second) symbol de-

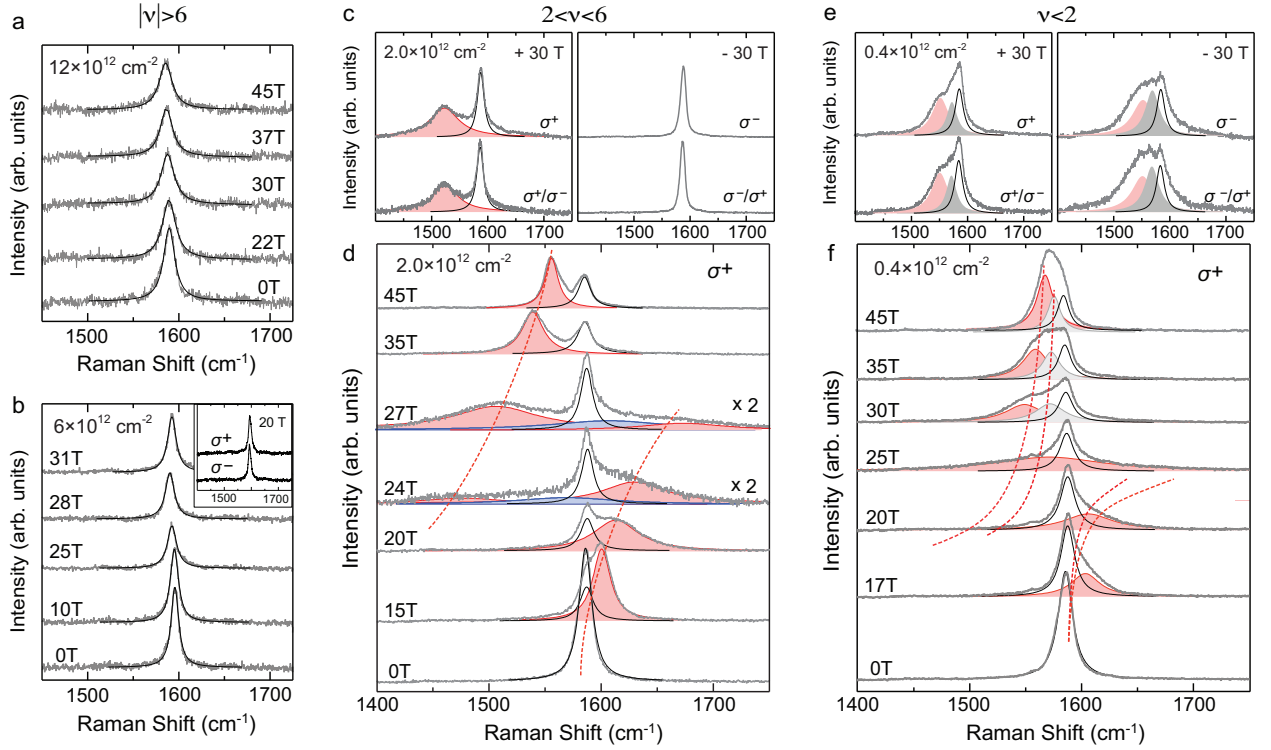


FIG. 2. (a,b) Unpolarized magneto-Raman spectra at carrier density corresponding to  $\nu \gtrsim 6$ . The inset compares 20T  $\sigma^+$  and  $\sigma^-$  circular polarized spectra. (c-f) Circular-polarized magneto-Raman spectra at (c,d)  $2 < \nu < 6$  and (e,f)  $\nu < 2$  for  $B > 15$ T. The dashed lines in (d,f) are guides to the eye for the B dependence of electron-phonon coupled modes at the  $(0 \rightarrow 1)$  MPR anticrossing. The blue curves in (d) are additional spectral components observed in the close vicinity of the intersection between a  $(0 \rightarrow 1)$  magneto-exciton and the  $E_{2g}$  phonon energy

finest the polarization of the excitation (scattered) light. The polarization efficiency, i.e. the ratio of light transmitted through two circular polarizers with parallel or crossed helicities, is  $\sim 90\%$  for the incident light, while it is  $\sim 75\%$  for the scattered light in the spectral range of interest  $\sim 1450$ - $1700 \text{ cm}^{-1}$ . The resolution is  $\sim 1.9 \text{ cm}^{-1}$ .

Figs. 2, 3 show the polarization- and B dependence of the G peak at different carrier densities. The Raman spectra can be classified in three categories, as a function of  $\nu$ . At high carrier density, corresponding to  $|\nu| \geq 6$ , the G peak does not reveal MPR-induced splitting nor any polarization dependence, Figs. 2a,b. As the filling factor decreases ( $\nu < 6$ ), a significant change in the spectra is observed. Figs. 2c,d plot spectra measured on the sample annealed, degassed and kept in  $< 10^{-4}$  mbar vacuum. The G peak exhibits a strong, anticrossing-like, splitting reaching  $\sim 150 \text{ cm}^{-1}$  ( $\sim 20 \text{ meV}$ ) for  $B = 25 \text{ T}$ . The electron-phonon coupled modes appear only in the  $\sigma^+$  and  $\sigma^+/\sigma^-$  geometry, while the G peak does not split nor shift in  $\sigma^-$  and  $\sigma^-/\sigma^+$  polarizations. This indicates n-type doping with estimated carrier density  $\sim 2.0 \times 10^{12} \text{ cm}^{-2}$ . The observed behavior reveals the MPR at  $2 < \nu < 6$ . Indeed, the condition  $\nu = 6$  is met at  $B \sim 10 \text{ T}$ , with a coupling of  $0 \rightarrow 1$  magnetoexcitons and  $E_{2g}$  phonons at higher B. As the carrier density further

decreases, so that  $\nu < 2$ , the G peak splitting changes (Figs. 2e,f). In contrast to the  $2 < \nu < 6$  case, the coupled modes now appear in both  $\sigma^+$  and  $\sigma^-$  polarizations. Moreover, the spectra measured at  $B \geq 30 \text{ T}$  reveal that the coupled-mode consists of two Lorentzians. Their relative intensities depend on the helicity of the incoming light. We assign these two components to the  $\sigma^\pm$  polarized modes originating from the coupling of  $E_{2g}$  phonons with  $0 \rightarrow 1$  and  $-1 \rightarrow 0$  magnetoexcitons. We assign the fact that  $\sigma^\pm$  polarized modes do not completely disappear in the measurements with opposite helicity to the imperfect polarization selectivity of our setup, combined with sample inhomogeneity, as discussed below.

For the strongest anticrossing, at  $B_{MRP}^0$ , the observed behavior can be described using the resonance approximation formula [15] for pairs of strongly coupled  $\Omega_\pm^\sigma$  circularly polarized modes:

$$\Omega_\pm^\sigma = \frac{\Omega_\Gamma + \Omega_0}{2} \pm \sqrt{\left(\frac{\Omega_\Gamma - \Omega_0}{2}\right)^2 + \frac{g_\sigma^2}{\hbar^2}}, \quad (1)$$

$$g_{\sigma^+}(2 < \nu < 6) = g_0 \frac{a}{l_B} \sqrt{6 - \nu}, \quad g_{\sigma^-}(2 < \nu < 6) = 0,$$

$$g_{\sigma^\pm}(0 < \nu < 2) = g_0 \frac{a}{l_B} \sqrt{2 \pm \nu}, \quad g_0 = \sqrt{\frac{3}{2}} \sqrt{\frac{\lambda_\Gamma}{4\pi}} \gamma_0.$$

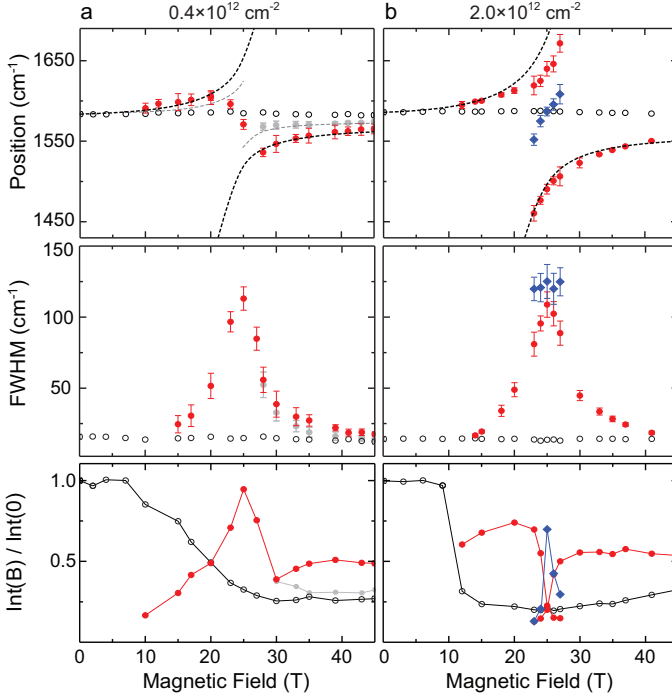


FIG. 3. Peak position, FWHM and normalized intensity of the G-peak Lorenzian components as a function of magnetic field. Open black circles represent the central  $E_{2g}$  phonon line component. Filled red and grey circles are the electron-phonon coupled modes. Dashed lines plot Eqs.1. Blue squares show additional components observed in close vicinity of the resonance  $B_{MPR}^0 \approx 25T$

Here,  $\Omega_{\pm}^{\sigma}$  is the frequency of the upper (+) or lower (-) coupled mode,  $\Omega_{\Gamma} \sim 1581\text{cm}^{-1}$  is the bare  $E_{2g}$  phonon energy,  $\Omega_0 = \sqrt{2}\hbar\tilde{c}/l_B$ ,  $a$  ( $= 2.46\text{\AA}$ ) is the graphene lattice constant.  $g_0$  describes the EPC at zero B in terms of the nearest neighbor hopping integral  $\gamma_0$ , and the dimensionless coupling constant  $\lambda_{\Gamma}$ [7, 13].

The set of equations (1) describe the energies of the two branches of mixed phonon and magneto-exciton modes for an idealized non-strained, homogeneously doped SLG, also neglecting broadening of the bare excitations due to inelastic and elastic scattering. The detailed description of MPR and its manifestation in the Raman spectrum of realistic samples should include broadening of the optical phonon,  $\gamma_{\Gamma}$ , and of the magneto-exciton,  $\gamma_e$ , as well as possible inhomogeneity of doping and fluctuations of local strain. In a sample with a spatially varying carrier density,  $g^{\sigma+}$  and  $g^{\sigma-}$  become coordinate-dependent, which would lead to inhomogeneous broadening of the MPR fine structure. Inhomogeneous strain in SLG generates a pseudo-magnetic field,  $B_{str}$ , with opposite sign in the K and K' valleys[5, 27, 28], giving a total field  $B_K = B + B_{str}$  at **K** and  $B_{K'} = B - B_{str}$  at **K'**. In strained graphene nanobubbles, it was reported that  $B_{str}$  can reach 200-400T[29]. In nominally unstrained SLG, the residual strain caused by nm-scale height variations

and defects can induce random  $B_{str} > 10T$ [30]. Locally, this splits a simple anticrossing into mixing and splitting of each  $\sigma^{\pm}$ -polarized phonon and two magneto-excitons, now distinguished by different LL energies in the two valleys, leading to an even more complicated fine structure. Also, the quantum efficiency (i.e. the probability of a scattering event per incoming photon) of  $0 \rightarrow 1$  and  $-1 \rightarrow 0$  magneto-excitons in Raman scattering[22] is much lower than the quantum efficiency of the  $\Gamma$ -point optical phonon, manifested in the G peak area,  $A(G)$ [31].

All these factors can be taken into account in the formula for the spectral weight of Raman scattering at MPR derived using the random phase approximation[32]:

$$I^{\sigma^{\pm}}(\omega) = \frac{A(G)}{2\pi} \left\langle \text{Im} \frac{1}{\omega - \Omega_G - i\gamma_{\Gamma} \mp \frac{\lambda_{\Gamma}}{4\pi} \sum_{\alpha} \frac{\Omega_{0,\alpha}^2}{\omega - \Omega_{0,\alpha} - i\gamma_e}} \right\rangle, \quad (2)$$

where angle brackets stand for the averaging over the spatial fluctuations of a strain-induced  $B_{str}$  with zero average, and over fluctuations of electron density. These together determine the local values of the inter-LL separation,  $\Omega_{0,\alpha} = \sqrt{2}\hbar\tilde{c}/l_{B_{\alpha}}$ , and the local filling factors of the  $n = 0$  and  $n = \pm 1$  LLs in each of the two valleys  $\alpha = K, K'$ . Eq.(1) can also be obtained from Eq.(2) neglecting strain and carrier density fluctuations.

To measure the EPC, we fit Eq.(1) to the data of Fig.3. The high-field anticrossing branches ( $\Omega_{-}$ ) are most accurately resolved and therefore most suitable for fitting. The energy of the unperturbed electronic transition  $\Omega_0$  is calculated using  $\tilde{c} = 1.08 \times 10^6\text{m/s}$ , which corresponds to the MPR at 25T. For low carrier density ( $0.4 \times 10^{12}\text{cm}^{-2}$ ), we obtain  $g_0^{\sigma+} = 17\text{cm}^{-1}/T^{1/2}$  and  $g_0^{\sigma-} = 12\text{cm}^{-1}/T^{1/2}$ . For intermediate carrier density ( $2.0 \times 10^{12}\text{cm}^{-2}$ ), fitting yields  $g_0^{\sigma+} = 21\text{cm}^{-1}/T^{1/2}$ . Plugging the fitted  $g^{\sigma\pm}$  into Eq.(1), we find  $\nu$  at the resonance B, and extract a carrier density  $\sim 0.4 \times 10^{12}$  and  $\sim 1.6 \times 10^{12}\text{cm}^{-2}$  for low and intermediate density samples, respectively, in excellent agreement with those derived from the Raman spectra. Taken together, these results allow us to deduce the dimensionless EPC. Notably, the  $\lambda_{\Gamma}$  we obtain for the low and intermediate density samples differ by less than 5%. We get  $\lambda_{\Gamma} \approx 0.028$ , in remarkable agreement with that measured from  $\text{FWHM}(G)$  in undoped samples at  $B=0$ , and predicted by density functional theory[3, 7, 13].

Finally, we discuss the observation of Raman scattering in the middle of the MPR anticrossing gap. The spectra for the intermediate carrier density show an additional component, indicated in blue in Fig.2d. This is detected over a narrow B range, between 23 and 27T, reaching maximum intensity at 25T. Though we are not able to spectrally resolve this component at lower carrier densities, a similar sharp increase of intensity is observed in the vicinity of the MPR at 25T (Fig.3a, bottom panel). Eq.(2) shows that a finite Raman scattering inside the anticrossing gap is a specific signature of MPR in strained



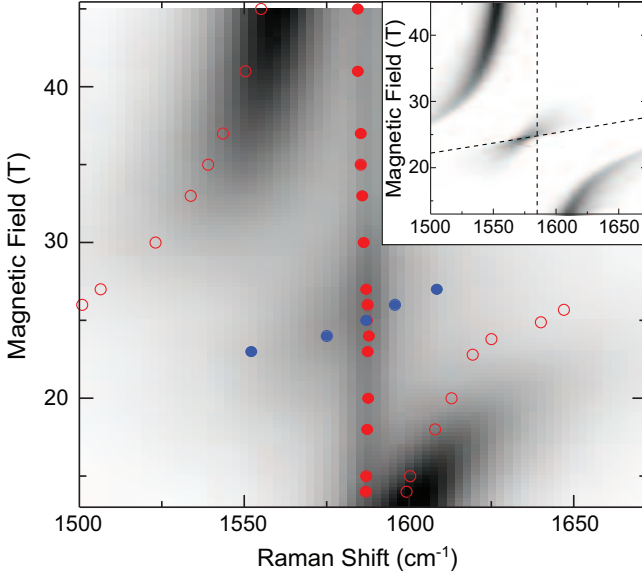


FIG. 4. MPR in SLG with  $\langle \rho_s \rangle = 1.6 \times 10^{12} \text{cm}^{-2}$  in the presence of inhomogeneous strain,  $\langle B_{str}^2 \rangle^{1/2} = 6 \text{T}$ , calculated using Eq.(2) with  $\Omega_L \sim 1581 \text{cm}^{-1}$ ,  $\gamma_L \approx 12.3 \text{cm}^{-1}$ ,  $\gamma_e = 50 \text{cm}^{-1}$ , and  $\langle \delta \rho_s^2 \rangle^{1/2} = 3 \times 10^{11} \text{cm}^{-2}$ . Symbols are experimental results. The inset shows the calculated Raman intensity, neglecting LL broadening, depolarisation, inhomogeneity, but with strain-induced inhomogeneous  $B_{str}$ . Dashed lines indicate the unperturbed  $E_{2g}$  phonons and (0,1) magneto-excitons

SLG. Fig.4 plots the Raman intensity calculated for intermediate carrier density. To model the experiment, we assume density fluctuations described by a normal distribution with a  $\langle \rho_s \rangle \sim 1.6 \times 10^{12} \text{cm}^{-2}$  average and 20% standard deviation, imperfect light polarization leading to a 75%/25% mixture of  $\sigma^+$  and  $\sigma^-$  spectra, LL and  $E_{2g}$  phonon broadening of  $\gamma_e = 50 \text{cm}^{-1}$  and  $\gamma_L = 12 \text{cm}^{-1}$  and, most importantly,  $\langle B_{str}^2 \rangle^{1/2} = 6 \text{T}$ . Away from the  $\Omega_0 = \Omega_L$  MPR intersection, the Raman intensity follows the expected anticrossing MPR behavior, while the increased scattering in the middle of the anticrossing gap is due to the overall effect of inhomogeneous strain. This is further illustrated in the inset in Fig.4, with a Raman intensity map calculated for the ideal case of uniformly doped SLG ( $\delta \rho_s = 0$ ) exposed to the same random  $B_{str}$ , neglecting broadening ( $\gamma_L = \gamma_e = 0$ ) and depolarization.

In summary, we used polarization-resolved high-field magneto-Raman to investigate magneto-phonon resonances in graphene. By varying the filling factor, we identified different types of G-peak magnetic-field dependencies, providing a comprehensive experimental evidence of MPR on circularly polarized phonons. We also detected

an unexpected increase of Raman intensity in the middle of the MPR anticrossing gap and assigned it to mixing and splitting of electron-phonon coupled modes caused by fluctuations of strain-induced pseudo-magnetic fields.

We acknowledge funding by NHMFL UCGP-5068, DOE/BES DE-FG02-07ER46451, DOE/BES DE-FG02-06ER46308, the Robert A. Welch Foundation C-1509, EPSRC grants GR/S97613/01, EP/E500935/1, ERC grant NANOPOTS, EU grants GENIUS and RODIN, two Royal Society Wolfson Research Merit Awards, NSF ECCS 0925988, ERC AdG 'Graphene and Beyond', the NSF Cooperative Agreement No. DMR-0654118, the State of Florida, and the DOE.

\* smirnov@magnet.fsu.edu

- [1] V.L. Gurevich and Yu.A. Firsov, Zh. Eksp. & Teor. Fiz. **40**, 199 [Sov. Phys. JETP **13**, 137] (1961).
- [2] Yu. A. Firsov et al. in *Landau Level Spectroscopy*, edited by G. Landwehr and E. I. Rashba (Elsevier, Amsterdam, 1991, p. 1192; R. J. Nicholas, *ibid.*, p. 779).
- [3] S. Piscanec et al. Phys. Rev. Lett. **93**, 185503 (2004)
- [4] A. C. Ferrari et al. Phys. Rev. Lett. **97**, 187401 (2006)
- [5] T. Ando, J. Phys. Soc. Jpn. **75**, 124701 (2006)
- [6] A. C. Ferrari, Solid State Commun. **143**, 47 (2007)
- [7] D. M. Basko et al. Phys. Rev. B. **80**, 165413 (2009).
- [8] S. Pisana et al. Nature Mater. **6**, 198 (2007)
- [9] J. Yan et al. Phys. Rev. Lett. **98**, 166802 (2007)
- [10] A. Das et al. Nature Nanotech. **3**, 210 (2008)
- [11] A. Das et al. Phys. Rev. B **79**, 155417 (2009)
- [12] J. Yan et al. Phys. Rev. B **80**, 241417(R) (2009)
- [13] M. Lazzeri et al. Phys. Rev. B **73**, 155426 (2006).
- [14] T. Ando, J. Phys. Soc. Jpn. **76**, 024712 (2007).
- [15] M. O. Goerbig et al. Phys. Rev. Lett. **99**, 087402 (2007).
- [16] T. Ando, J. Phys. Soc. Jpn. **76**, 104711 (2007).
- [17] J. Yan et al. Phys. Rev. Lett. **105**, 227401 (2010).
- [18] C. Faugeras et al. Phys. Rev. Lett. **107**, 036807 (2011).
- [19] C. Faugeras et al. Phys. Rev. Lett. **103**, 186803 (2009).
- [20] Y. Kim *et al.*, Phys. Rev. B **85**, 121403(R) (2012).
- [21] A.H. Castro Neto *et al.*, Rev. Mod. Phys. **81**, 109 (2009).
- [22] O. Kashuba, V. I. Fal'ko, Phys. Rev. B **80**, 241404 (2009).
- [23] X. Li et al. Science **324**, 1312 (2009)
- [24] S. Bae et al. Nature Nanotech. **5**, 574 (2010)
- [25] L.G. Cancado et al. Nano Lett. **11**, 3190 (2011)
- [26] A. Das *et al.*, Nature Nanotechnology **3**, 210 (2008).
- [27] S. V. Iordanskii, A. E. Koshelev, JETP Lett. **41**, 574 (1985).
- [28] A. F. Morpurgo, F. Guinea, Phys. Rev. Lett. **97**, 196804 (2006).
- [29] N. Levy *et al.*, Science **329**, 544 (2010).
- [30] N.-C. Yeh *et al.*, Surf. Sci. **605**, 1649 (2011).
- [31] D. M. Basko, New Journal of Physics **11**, 095011 (2009).
- [32] O. Kashuba and V. I. Fal'ko, New J. Phys. **14**, 105016 (2012)